Saharan dust storms and indirect aerosol effects on clouds: CRYSTAL-FACE results

Kenneth Sassen,[1] Paul J. DeMott,[2] Joseph M. Prospero,[3] and Michael R. Poellot[4]

Received 19 March 2003; revised 8 May 2003; accepted 15 May 2003; published 24 June 2003.

[1] A recent field experiment in southern Florida using aircraft and polarization lidar shows that mineral dust particles transported from Saharan Africa are effective ice nuclei, apparently capable of glaciating a mildly supercooled ($-5.2°$ to $-8.8°$C) altocumulus cloud. These results are similar to those from Asian dust storm particles observed over the western US, suggesting that in the northern hemisphere major dust storms play a role in modulating climate through the indirect aerosol effect on cloud properties. If this is true of desert dusts in general, then even minor aeolian emissions could have an effect on regional weather and climate. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 3360 Meteorology and Atmospheric Dynamics: Remote sensing. **Citation:** Sassen, K., P. J. DeMott, J. M. Prospero, and M. R. Poellot, Saharan dust storms and indirect aerosol effects on clouds: CRYSTAL-FACE results, *Geophys. Res. Lett.*, *30*(12), 1633, doi:10.1029/2003GL017371, 2003.

1. Introduction

[2] Yearly cycles of dust storms from the Earth's major desert regions are a well-known phenomenon caused by the interaction of seasonal weather patterns and a fetch of surface deposits of dry mineral dust. Such events, however, are garnering increased scientific scrutiny because of recent developments. A combination of increasingly sophisticated remote sensing instruments from the ground and Earth orbit, in situ aerosol probes, and long-range aerosol transport models are greatly facilitating the observation and study of these dust clouds [*Kaufman et al.*, 2002]. In particular, the near real-time dissemination of satellite imagery, lidar network data, and transport model predictions at a number of international web sites is promoting this research. Extensive aerosol layers directly affect the radiative balance of the Earth-atmosphere system through scattering and absorption, thus increasing the local solar albedo (potentially cooling the surface) and heating the atmosphere, and indirectly by modifying cloud particle phase and size distribution. The indirect effect of aerosols on climate is highly uncertain [*IPCC*, 2001]. Such changes have been demonstrated in only a few cases because of the difficulty of obtaining conclusive evidence in the face of often extreme natural cloud variability. In view of the variety of climate change issues confronting scientists, an overarching question is whether human-induced land use changes leading to desertification, or climate change itself [*Liu et al.*, 2002], is increasing the occurrence and severity of dust storms in vulnerable areas of the globe.

[3] The two major source regions of dust storms, the immense deserts of the Mongolian and Saharan regions, have long been known to be capable of spreading debris over long distances [*Husar et al.*, 1997]. The evidence for indirect aerosol effects on clouds currently seems clear for the case of Asian dust. Early ice nuclei (IN) measurements in Japan [*Isono et al.*, 1959] established that advected Asian dust particles composed of clay were especially active IN, and could even represent an important world-wide source of these special particles. It was not until recently, however, that polarization lidar data yielded evidence that Asian dust was linked to changes in basic cloud properties [*Sassen*, 2002]. The lidar backscatter depolarization technique [*Sassen*, 2000] is well suited to identifying Asian dust because the relatively large (i.e., of a size comparable to the wavelength), sharp-edged crystalline particles [*Okada et al.*, 2001] generate strong depolarization, even approaching that of some cirrus clouds. Cirrus have been associated with Asian dust layers over Japan [*Murayama et al.*, 2001] and as far away as the Great Basin of the US [*Sassen*, 2002], suggesting the dust helped initiate ice cloud formation. Moreover, in comparison to the properties of a 10-y sample of midlatitude cirrus, case studies of cirrus connected to Asian dust layers were found to be considerably warmer in temperature, suggesting cloud phase changes due to the IN activity of the aerosol [*Sassen*, 2002].

[4] Recently, data collected during the Cirrus Regional Study of Tropical Anvils and Cirrus Layers-Florida Area Cirrus Experiment (CRYSTAL-FACE) has shed light on the potential cloud-altering properties of transported Saharan dust. Although the main purpose of this intensive aircraft and remote sensing experiment was to research subtropical thunderstorms and the cirrus derived from their anvils, it was recognized that Saharan dust clouds, which during summer are regularly transported across the Mid-Atlantic Ocean to the Caribbean and Southern Florida [*Prospero*, 1999], were likely to be encountered. The data given here and in *DeMott et al.* [2003] support the deduction that African dust particles have properties similar to their Asian counterparts.

2. Remote Sensing and In Situ Datasets

[5] During the month of July 2002, a variety of instruments participated in the CRYSTAL-FACE field campaign in

[1]Geophysical Institute, Univ. of Alaska Fairbanks, Fairbanks, AK, USA.
[2]Dept. of Atmospheric Sciences, Colorado State Univ., Fort Collins, CO, USA.
[3]Rosenstiel School of Marine and Atmospheric Science, Univ. of Miami, FL, USA.
[4]Atmospheric Sciences Department, Univ. of North Dakota, Grand Forks, ND, USA.

0094-8276/03/2003GL017371$05.00

and around southern Florida. Of interest here are the Colorado State Univ. continuous flow diffusion chamber (CFDC) [*Rogers et al.*, 2001] aboard the Univ. of North Dakota Citation aircraft, and the Facility for Atmospheric Remote Sensing (FARS) Polarization Diversity Lidar (PDL) [*Sassen et al.*, 2001]. The CFDC on this occasion was operated in a mode that allowed ice nucleation only through the heterogeneous mechanism: The temperature and humidity conditions used for aerosol processing prohibited homogeneous nucleation [*DeMott et al.*, 2003]. Thus these data represent the heterogeneous IN concentrations active at a temperature of -36 ± 2°C. The Citation IN data used here were obtained on descent into Key West, ~135 km to the south of the lidar site and ~1-h after the end of PDL data collection.

[6] The mobile PDL was deployed at the Ochopee site in the western Everglades. This fully scannable lidar is a testbed for high-resolution (1.5-m by 0.1-s), dual-wavelength (1.06 and 0.532 μm) polarization lidar techniques. As configured in CRYSTAL-FACE, linear depolarization ratios (δ, the ratio of the laser powers returned in the orthogonal-to-parallel polarization channels) were obtained at both wavelengths, although the more sensitive 0.532 μm data are used here to characterize the aerosol. Note that the δ values measured in aerosols represent a mixture of particulate and (nearly non-depolarizing) molecular backscattering at this wavelength. On 28 and 29 July, the final days of the campaign, interesting PDL data were obtained from elevated aerosol layers. Enhanced parallel-polarized aerosol backscattering (i.e., above the molecular signal) was noted extending well above the boundary layer, and the depolarized signals confirmed the presence of relatively large nonspherical particles.

[7] Figure 1 provides PDL range-normalized returned power and δ-value height-versus-time displays for a ~1-h period on the morning of 29 July. The observations were initially designed to study the cirrus layers, and in doing so captured a depolarizing aerosol layer extending from the top of the boundary layer (~1.7 km above mean sea level, MSL) to ~5.5 km. The peak $\delta = 0.10-0.15$ in the aerosol are lower than the maximum 0.25 values measured in elevated Asian aerosol layers by the PDL at FARS [*Sassen*, 2002], but considering the relatively stronger molecular backscattering in the Florida measurements, the amount of *aerosol* depolarization appears similar. In comparison, $\delta \approx 0$ are typical of the boundary layer (haze) aerosol, while higher $\delta \sim 0.2$ to 0.4 are found in the cirrus, except for the $\delta < 0.05$ sometimes present from horizontally oriented plate crystals in the upper cloud layer [see *Sassen and Benson*, 2001].

[8] Shown at the right side of Figure 1 is a shorter data file adjusted to capture the passage of a midlevel altocumulus cloud. Since the lidar system gain was decreased in order to capture the more strongly-scattering liquid cloud, the returns are too weak to calculate δ in air much above the boundary layer. However, it can be assumed that the aerosol conditions aloft remained similar because of the prior temporal consistency of aerosol returns. The altocumulus produces near-zero depolarization at the liquid cloud base from spherical droplets, with a gradual increase aloft due to multiple scattering effects in the optically denser portions of the cloud. Note that as the layer height decreases it temporally glaciates at 1516 UTC, leaving ice virga displaying $\delta \approx 0.3-0.4$ descending ~1.0 km below the cloud. This glaciation occurs close to the top of the dust layer. The

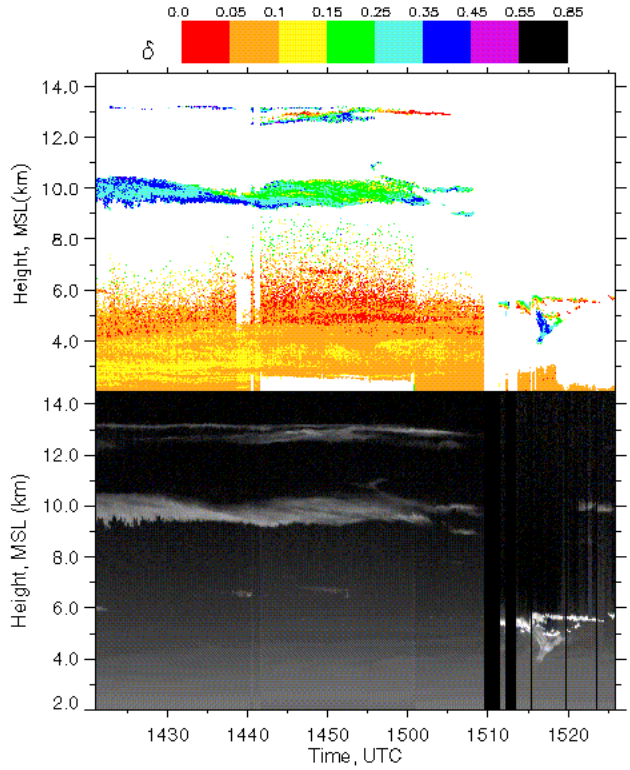

Figure 1. PDL linear depolarization ratio (see color δ scale at top) and relative returned power (in a logarithmic gray scale) height-versus-time displays on the morning of 29 July 2002, from the Ochopee field site of the CRYSTAL-FACE program. Note that low-altitude signals often cannot be used to calculate δ-values because of strong off-scale signals. Depicted are strongly depolarizing ($\delta \sim 0.2$ to 0.4) upper tropospheric cirrus clouds, aerosols ($\delta \sim 0.10$ to 0.15) extending up to ~5.5 km, and at far right a supercooled liquid altocumulus cloud ($\delta \approx 0$ at cloud base). Note the temporary glaciation of this cloud as it descended into the top of the dust layer.

average temperatures measured by the Citation during takeoff and landing at the altocumulus cloud base and top heights are a relatively warm −5.2° and −8.8°C, respectively. This can be interpreted in terms of an aerosol-induced cloud phase change, assuming that the aerosols mixed into the cloud are especially active IN. Note that there is no evidence for ice crystal precipitation from aloft that could have "seeded" the layer.

[9] Figure 2 gives later PDL data that is more contemporary with the Citation CFDC data. At left are 8-min average profiles of PDL backscattering and δ, and at right, in situ data gathered near Key West at around 2100 UTC using the Forward Scatter Spectrometer Probe (FSSP), which was used to sample cloud droplets with diameters larger than ~3.0 μm, and the CFDC. (Note that the FSSP analysis is designed for spherical particles with the refractive index of water, so that unknown sizing uncertainties can be expected for the aerosol.) Nonetheless, the FSSP detected a gradual increase in concentrations starting just below 4.0 km, followed by a strong increase at the top of the boundary layer at ~1.7 km, which is consistent with

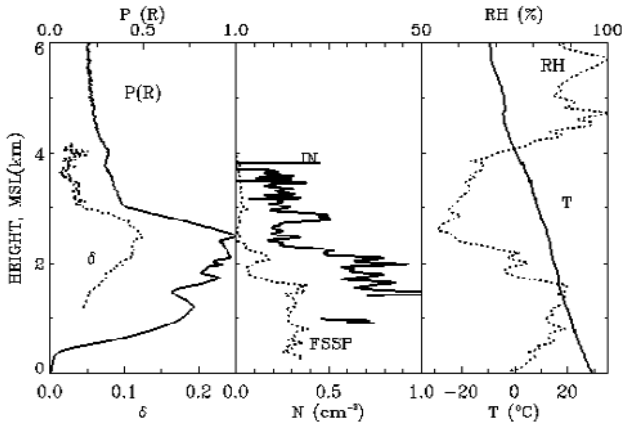

Figure 2. Comparison of in situ aerosol data collected by the Citation aircraft with 8-min (1941–1948 UTC) average PDL profiles of backscattered power P(R) and depolarization δ. Note the correlation between the IN concentrations, the FSSP counts (for particles >∼3 μm), and the lidar data (valid >1-km). The gap in IN data near 1.2 km is due to the blockage of the instrument intake to obtain a filter sample. The temperature T and relative humidity RH profiles at right are the average measured during Citation takeoff and landing near Key West, bracketing the lidar measurements.

hygroscopic aerosol swelling in the moist boundary layer. At the same time, IN concentrations are first evident at 3.8 km and gradually increase down to at least 0.8 km, where the device was turned off. The heterogeneous IN concentrations of up to 1.0 cm^{-3} are among the highest ever measured with the CFDC [*DeMott et al.*, 2003].

[10] This in situ evidence confirms the presence of large aerosols within the region the lidar shows contained supermicron sized nonspherical particles, and that these particles were unusually active IN. The average lidar profiles in Figure 2 can be expected to differ somewhat because of spatial and temporal variations in aerosol conditions, but indicate the top of strong aerosol backscattering at ∼3.0 km, with weaker aerosol contributions extending to just above 4.0 km. Such heights are typical of the top of the Saharan air layer over the western Atlantic [*Reid et al.*, 2002]. The strongest aerosol depolarization is generated from ∼2.0–3.0 km, but the δ decrease rapidly within the boundary layer down to a height of ∼1.2 km, where the data become unreliable due to the incomplete laser/receiver beam overlap. The $\delta < 0.05$ in the boundary layer may reflect the fact that the aerosol depolarization was overwhelmed by the local marine haze, although they too may have become activated in the humid boundary layer if coated with hygroscopic materials.

3. Aerosol Chemical Dataset

[11] The combined in situ and remote sensing data on this occasion, and the historical record, suggest an aerosol dominated by soil particles of African origin, with properties similar to those of Asian dust. To confirm this finding, ground-based aerosol samples collected as part of a long-term study at a coastal site in Miami [*Prospero*, 1999] have been analyzed. Daily filter samples are collected on Virginia Key, several kilometers east of mainland Miami, only during periods of on-shore winds to minimize impacts from local aerosol sources. Soluble components are extracted and analyzed for non-sea salt (nss) sulfate, nitrate, and Na (which is converted to equivalent sea-salt aerosol). The extracted filter is turned to ash at 500°C and the residue converted to an equivalent mineral dust concentration.

[12] Selected aerosol concentrations from early June through mid-August are shown in Figure 3. Some mineral dust was present during most of the month of July, but the maximum concentrations were observed on 28 and 29 July. The mean concentration, 16.4 μg m^{-3}, is nearly identical to the long-term (1989–1996) July mean of 16.3 μg m^{-3} [*Prospero*, 1999]. The July concentrations of nss-$SO_4^=$ are close to the mean of 1.84 μg m^{-3}. Sea-salt concentrations are substantial, averaging 8.0 μg m^{-3}(compared to the 7.57 μg m^{-3} mean).

4. Conclusions

[13] Our knowledge of the properties and potential climatic impact of major desert dust storm events is evolving rapidly. In addition to the recognized direct solar scattering effects and the indicated indirect effects on clouds, there appears to be a plethora of processes affected by the long-range transport of desert dusts. These include the effects on human health through air quality, scavenging of pollution, chemical reactivity affecting ozone concentrations, and the increased fertilization of oceans and arid lands. We have explored here the ability of Saharan dust to affect cloud phase because of their unusual ice particle nucleation properties: The resultant alternations in basic cloud properties and lifetimes may affect the radiation balance of the earth-atmosphere system.

[14] The data indicate that the properties of Saharan dust are similar to those from Asian deserts in terms of their potential for indirect cloud effects. The altocumulus glaciation observed in this study at the upper boundary of a transported African dust layer is analogous to findings reported in *Sassen* [2002] for Asian dust over the western US, and is strongly suggestive of a cloud seeding affect

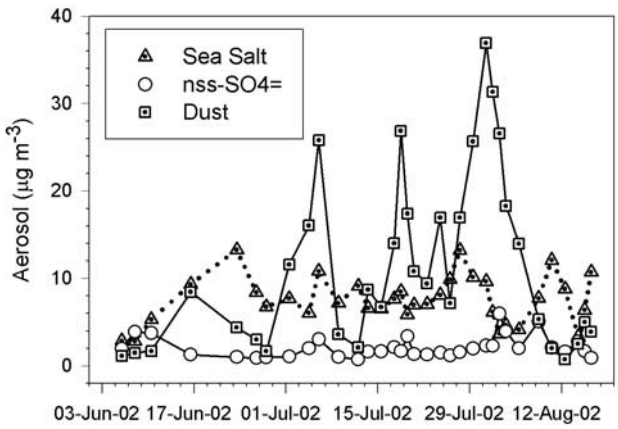

Figure 3. Comparison of Miami aerosol mass concentrations bracketing the July 2002 period of the CRYSTAL-FACE campaign. The samples where obtained on filters changed at mostly 1-day intervals.

from introduced IN. (Indeed, the observed cloud temperatures are only just suitable for artificial cloud seeding using silver iodide agents, which, like clay, display structural epitaxy with ice.) The cloud temperatures appear to be unusually warm, although *Hobbs and Rangno* [1985] found evidence for enhanced ice crystal concentrations and eventual cloud glaciation in some altocumulus with similar cloud top temperatures, which they attributed to contact nucleation during cloud-top mixing. This scenario may apply in our case, where the nature of the IN (likely introduced into cloud base) has been determined independently. It should also be pointed out that clay materials have been found to be uniquely active contact IN with median (750 μm-diameter) drop freezing temperatures of about −8°C for montmorillonite and −12°C for kaolinite [*Pitter and Pruppacher*, 1973]. Moreover, *Rosenfeld et al.* [2001] recently reported in situ data indicating ice nucleation in Saharan dust-contaminated clouds at as warm as −8°C.

[15] Since a large part of the eastern US [*Perry et al.*, 1997] and Atlantic coastal waters [*Arimoto et al.*, 1995] are impacted to some degree by African dust every summer, the processes that we observed in Florida could affect cloud properties over a large area of the US. It is also apparent that a portion of the deep convection intensively studied during the July 2002 CRYSTAL-FACE campaign could have been significantly affected by the rich source of African IN. These observations coupled with those of Asian dust over the western US suggest that mineral dust could have a significant impact on climate over much of the US despite the great transport distances involved. Importantly, it remains to be determined whether desert dusts in general behave in a similar fashion, such that more minor desert areas could have important regional effects on weather and climate processes.

[16] **Acknowledgments.** This research was supported by NASA grants NAG5-13418 and NAG5-11476 from the CRYSTAL-FACE program, and DOE grant DEFG0394ER747 from the Atmospheric Radiation Measurement Program for PDL development.

References

Arimoto, R., Trace elements in the atmosphere over the North Atlantic, *J. Geophys. Res.*, *100*, 1199–1213, 1995.

DeMott, P. J., K. Sassen, M. R. Poellot, D. Baumgardner, D. C. Rogers, S. Brooks, A. J. Prenni, and S. M. Kreidenweis, African dust aerosols as atmospheric ice nuclei, *Geophys. Res. Lett.*, *30*, 10.1029/2003GL017410, 2003.

Hobbs, P. V., and A. L. Rangno, Ice particle concentrations in clouds, *J. Atmos. Sci.*, *42*, 2523–2549, 1985.

Husar, R. B., J. M. Prospero, and L. L. Stowe, Characterization of tropospheric aerosols over the oceans with the NOAA advanced very high resolution radiometer optical thickness operational product, *J. Geophys. Res.*, *102*, 16,889–16,909, 1997.

IPCC, "Climate Change Science: An Analysis of Some Key Questions", National Academy Science [http://books.nap.edu/books/0309075742/html/], 2001.

Isono, K., M. Komabayasi, and A. Ono, The nature and origin of ice nuclei in the atmosphere, *J. Meteorol. Soc. Japan*, *37*, 211–233, 1959.

Liu, P., G. A. Meehl, and G. Wu, Multi-model trends in the Sahara induced by increasing CO_2, *Geophy. Res. Lett.*, *29*, 10.1029/2002GL015923, 2002.

Kaufman, Y. K., D. Tanré, and O. Boucher, A satellite view of aerosols in the climate system, *Nature*, *419*, 215–223, 2002.

Murayama, T. N., and coauthors, Ground-based network observation of Asian dust events of April 1998 in east Asia, *J. Geophys. Res.*, *106*, 18,345–18,360, 2001.

Okada, O., J. Heintzenberg, K. Kai, and Y. Qin, Shape of atmospheric mineral particles collected in three Chinese arid-regions, *Geophys. Res. Lett.*, *28*, 3123–3126, 2001.

Perry, K. D., T. A. Cahill, R. A. Eldred, D. D. Dutcher, and T. E. Gill, Long-range transport of North African dust to the eastern United States, *J. Geophys. Res.*, *102*, 11,225–11,238, 1997.

Pitter, R. L., and H. R. Pruppacher, A wind tunnel investigation of freezing of small water drops falling at terminal velocity in air, *Q. J. R. Meteorol. Soc.*, *99*, 540–550, 1973.

Prospero, J. M., Longterm measurements of the transport of African mineral dust to the Southestern United States: Implications for regional air quality, *J. Geophys. Res.*, *104*, 15,917–15,927, 1999.

Reid, J. S., and coauthors, Dust vertical distribution in the Caribbean during the Puerto Rico Dust Experiment, *Geophys. Res. Lett.*, 10.1029/2001GL014092, 2002.

Rogers, D. C., P. J. DeMott, S. M. Kreidenweis, and Y. Chen, A continuous flow diffusion chamber for airborne measurements of ice nuclei, *J. Atmos. Oceanic Tech.*, *18*, 725–741, 2001.

Rosenfeld, D., Y. Rudich, and R. Lahav, Desert dust suppressing precipitation: A possible desertification feedback loop, *Proc. Natl. Acad. Sci. USA*, *98*, 5975–5980, 2001.

Sassen, K., Lidar Backscatter Depolarization Technique for Cloud and Aerosol Research, in "*Light Scattering by Nonspherical Particles: Theory, Measurements, and Geophysical Applications*", edited by M. L. Mishchenko et al., Academic, 393–416, 2000.

Sassen, K., Indirect climate forcing over the western US from Asian dust storms, *Geophys. Res. Lett.*, *29*, 10.1029/2001GL014051, 2002.

Sassen, K., and S. Benson, A midlatitude cirrus cloud climatology from the Facility for Atmospheric Remote Sensing: II. Microphysical properties derived from lidar depolarization, *J. Atmos. Sci.*, *58*, 2103–2112, 2001.

Sassen, K., J. M. Comstock, Z. Wang, and G. G. Mace, Cloud and aerosol research capabilities at FARS: The Facility for Atmospheric Remote Sensing, *Bull. Am. Meteorol. Soc.*, *82*, 1119–1138, 2001.

K. Sassen, 903 Koyukuk Drive, Univ. of Alaska Fairbanks, Fairbanks, Alaska 99775, USA. (Ksassen@gi.alaska.edu)